NATURAL COPROCESSING OF COAL AND TAR

by

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ABSTRACT

This study examines optically the interaction between coal and coal-tar pitch in a self-burning coal setting. Three distinct zones, a thick, lower carbonization, a narrow, middle active combustion and also a narrow, upper oxidation zone were identified. The presence of basic instead of a mosaic anisotropy on the solids is indicative of weathering/oxidation prior to combustion. Air could circulate easily through open fractures within the uncompacted coal fragments, thus facilitating combustion at depth. The coal was carbonized at approximately 500-550°C at a rate of heating of 10°C/min. and subsequently combusted at 550-600°C. The coal has also been thermally altered to produce solid (semi-coke/coke), liquid (tar) and volatile matter. Different types of tar have been observed microscopically, interacting with coal fragments by briquetting them and acting as hydrogen donors to vitrinite.

Because additives such as coal-tar pitch often modify and change the optical texture and nature of residues during laboratory processes such as coal hydrogenation and co-carbonization, useful information can be gained by studying the natural coprocessing of coal with coal-tar pitch using optical microscopy and petrographic techniques.

INTRODUCTION

There have been numerous studies on the interaction between coals of different rank and additives containing oxygen, nitrogen, sulphur, aromatic compounds, petroleum pitches and coal-tar pitches (Marsh, 1973; Marsh et al., 1973a,b,c; Marsh et al., 1974; Marsh et al., 1980; Marsh and Neavel, 1980; Mochida et al., 1979a,b,c), and between coal macerals (Goodarzi, 1984). These additives often modify and change the optical texture and nature of residues during such processes as liquefaction (hydrogenation) and co-carbonization of coal macerals (Shibaoka et al., 1980; Goodarzi, 1984).

It has been shown by Shibaoka et al., (1980) and later by Steller (1981) that during hydrogenation the interaction between a coal and a hydrogen-rich additive results in the formation of reaction rims termed 'hydrogenation rims' These rims are possibly the precursors of vitroplast because they form from the interaction of additive with the vitrinite particles in the coal and are rich in hydrogen. More recently, Gentzis and Goodarzi (1989) observed reaction rims formed around vitrinite particles due to the influence of carbonization by-products such as tar, in a self-burning coal wastepile in Coleman, Alberta, Canada.

Goodarzi (1984) reported similar reaction rims when highly reactive sporinite was co-carbonized with less reactive vitrinite in coals of the same rank. The reaction rims formed at the boundaries between carbonized vitrinite and sporinite had a distinct optical texture (size and anisotropy of the mosaic units). Goodarzi (1984) used coals from two different ranks (high-volatile bituminous C - % Ro = 0.62, and high-volatile bituminous A - % Ro = 1.04) and observed that the higher the rank of coal, the wider were the reaction rims. When the blend having the lower-reflecting vitrinite was carbonized, two types of optical texture were produced, an isotropic for vitrinite and a granular anisotropic for sporinite.

Three types of interactions may occur between a coal and an additive in a blend during carbonization which may result in the formation of different fluid phases. These are: fluid mixing, solvation and solvolysis (Mochida et al., 1979). During fluid mixing, the two miscible fluids forming from coal and additive create a new fluid with a new molecular composition and properties (Mochida et al., 1979). Solvation is considered to be more of a physical rather than a chemical process and involves the extraction or leaching of the coal by the additive, followed by a subsequent stabilization of the formed molecules in the fluid pitch additive. Solvolysis is a chemical process which involves simultaneous depolymerization and interaction of the coal and additive involving hydrogen transfer mechanism (Mochida et al., 1979).

This study deals with the interaction of organic components such as coal macerals and semicoke/coke fragments with tar generated by the self-burning of coal.

EXPERIMENTAL

A 440 cm deep channel was dug on the top of a self-burning coal wastepile at Coleman Collieries in Alberta and samples were taken from the oxidation (ash), combustion and carbonization zones as well as from unaltered coal. The hot samples were cooled in water immediately to prevent further oxidation and combustion.

The samples were then dried and crushed to pass -20 mesh (<850μm). They were subsequently polished and their maximum and minimum reflectances in oil (n_{oil} = 1.518) were measured using a Zeiss MPM II microscope, fitted with a Zonax microcomputer. Photomicrographs were taken under both plane-polarized light and with partially crossed polars.

RESULTS AND DISCUSSION

Gentzis and Goodarzi (1989) examined the organic petrology of the self-burning coal wastepile in Coleman, Alberta, Canada, (Figure 1a) and reported the reflectance profile of the different zones identified. These zones are: oxidation (ash), combustion and carbonization, all observed from top to base of the wastepile but their boundaries are not well defined (Figure 1b).

Carbonization zone

This zone is thick (~300 cm) and is subdivided into three subzones. The first subzone (B) is approximately 60 cm thick and consists mainly of warm, angular coal fragments. Two types of tar are observed (Figure 1c), an isotropic (% Romax = 1.07) and an anisotropic (% Romax = 2.35).

Subzone C is 180 cm thick and contains loose particles of hot, angular coal and semicoke/coke, becoming increasingly tarry towards the upper part. Three tar types are identified (Figure 1c), one is isotropic (% Romax = 0.72) and the other two are anisotropic (% Romax = 1.25 and 1.52 respectively) showing 'flow' texture.

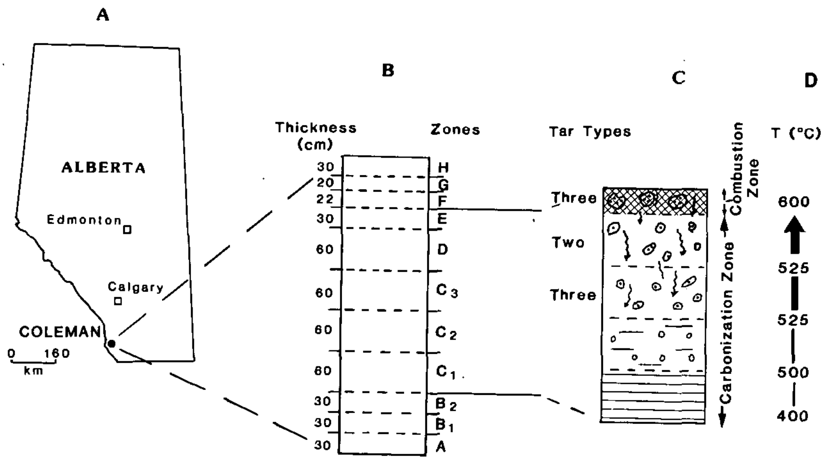

Figure 1 Location map of the coal wastepile in Coleman, Alberta (A), vertical profile of the self-burning coal wastepile showing different zones (B), types of tar (C) and temperature of their formation (D).

Subzone D is 60 cm thick consisting of angular semicoke and two tar types are present (Figure 1c). The isotropic tar has an Romax of 1.10%, while the anisotropic has an Romax of 1.66%.

Combustion zone

This zone is 30 cm thick and consists of a mixture of coke and ash. Both isotropic and anisotropic tars are present. The isotropic ones have an Romax of 0.86 and 1.13%, while the anisotropic ones have Romax values of 1.48, 1.90, 2.62 and 3.65% respectively. The high reflectance tar also shows granularity.

Oxidation (Ash) zone

The ash zone is 22 cm thick and consists of red ash and vents. Four types of tar are present, one isotropic (% Romax = 1.07) and three anisotropic (1.33, 1.53 and 1.77% Ro max respectively).

Heat generated by the combustion of coal alters the coal fragments in the oxidation zone and carbonization zone (Figure 1c) and produces solid residue (semicoke/coke), liquid (tar) and gases. The tar is mainly liquid to gaseous at the temperature of generation (500-550°C) but solidifies upon cooling and forms a pitch-like solid. The presence of the ash zone above the combustion zone in Coleman prevents the complete elimination of volatile by-products and, as a result, volatile matter escapes from vents developed at the side of the wastepile. These vents are impregnated by coal tar-pitch and often heat-affected fragments (semicoke/coke) are briquetted by tar (Gentzis and Goodarzi, 1989). Therefore, tar which formed due to coal devolatilization in the carbonization zone, such as in subzone C, migrates downwards in the wastepile due to force of gravity, penetrates the space among coal fragments and precipitates as isotropic to anisotropic by-products. The downward movement of tar is also due to a heat barrier produced by the combustion zone above (Figure 1b).

Similar observations were made by Goodarzi et al., (1988) on a partially combusted and coked bituminous coal seam from Aldridge Creek, British Columbia, Canada. A pitch-like, viscous material, which was soluble in chloroform-ethanol (87/13) azeotrope (similar to coal tar-pitch) formed from the carbonization of a medium-volatile bituminous coal seam (% Romax = 1.1), had migrated downwards filling the devolatilization vacuoles of the semicoke.

Figure 1c shows the various types of tar and their location in the wastepile and Figure 1d shows the temperature of their formation. Gentzis and Goodarzi, (1989) estimated that the coal was carbonized at a temperature of 500-550°C at a rate of heating of 10°C/min. and subsequently combusted at a higher temperature (550-600°C). The tar which formed in the semicoke subzone C_3 (~525°C) is isotropic, soft, typical of coal tar-pitch (Plate 1a). The coal rank in the Coleman section is medium-volatile bituminous (% Romax = 1.07), similar to the coal rank in Aldridge Creek. In both cases, coal was transformed to coke.

In Aldridge Creek, the coal seam is burning underground and under a sedimentary cover. As a result, the coal seam is directly transformed into carbonized residue. Therefore, no briquetting occurs but only impregnation of semicoke by tar takes place. In Coleman, coal is in fragmented form prior to burning, and is briquetted by tar-pitch as a result of self-burning. Most of the coke fragments in Coleman are subangular showing basic anisotropy, an indication that coal was probably weathered before being combusted (Goodarzi, et al., 1975).

Often, one or two waves of tar are observed filling cavities among semicoke fragments (Plate 1b). These two waves (types) of tar can be recognized on the basis of optical texture, an anisotropic, higher-reflecting tar which was obviously deposited initially, followed by an isotropic, lower-reflecting secondary tar.

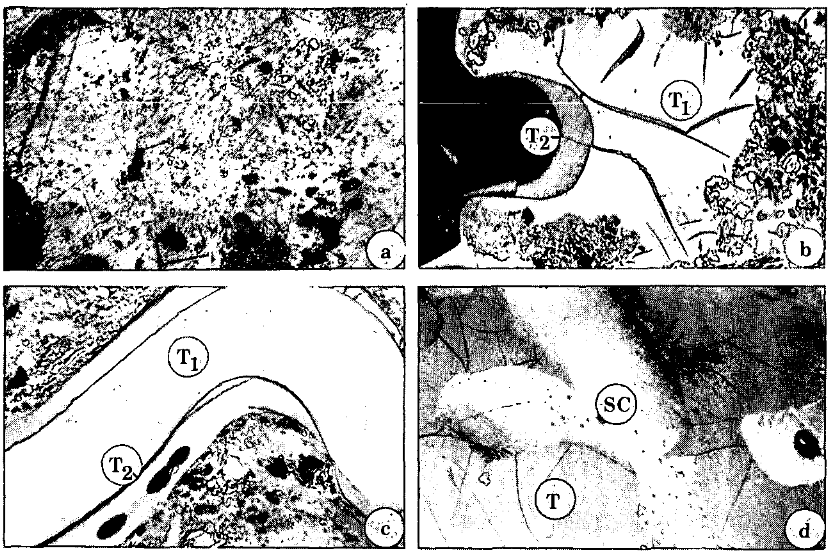

PLATE 1

All photomicrographs taken in black and white, plane-polarized light, under oil immersion. Long axis is 240μm.
a) Isotropic and soft coal tar-pitch, semicoke subzone (C_3); b) Two waves of tar filling cavity in semicoke subzone (C_3). The initial tar (T_1) is higher reflecting, whereas the secondary tar (T_2) is isotropic; c) Two generations of tar showing fluidity; the older one (T_1) has a granular morphology and higher reflectance, the younger one (T_2) shows devolatilization vacuoles and lower reflectance; d) Semicoke fragments (SC) showing devolatilization vacuoles being surrounded by tar (T).

Tar also shows evidence of fluidity, an indication of its mobility. Plate 1c shows two types of tar, a higher-reflecting, slightly granular tar deposited initially and a secondary, lower-reflecting tar exhibiting devolatilization vacuoles. Both tars have infilled the crack between the semicoke fragments and have taken its shape. Occasionally, the subangular fragments of semicoke show devolatilization vacuoles and are completely surrounded by tar (Plate 1d). Within the semicoke subzone (C_3), mixing of semicoke and coal fragments may take place (Plate 2a-b) with tar binding both fragments. The low-reflecting tar is hydrogen-rich and apparently is able to react with the coal particles producing a distinct reaction rim (Plate 2a). This did not occur between the tar and semicoke fragments which show granular mosaic texture but no apparent reaction rim (Plate 2b).

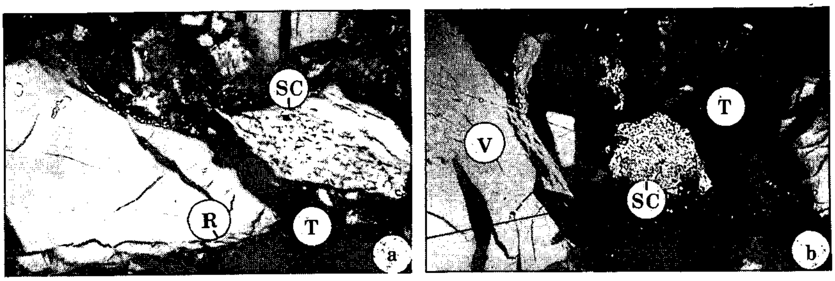

PLATE 2 Conditions same as Plate 1.

a) Low-reflecting, hydrogen-rich tar (T) reacting with coal and semicoke (SC) fragments. Note the presence of a reaction rim (R) between coal and tar; b) Similar to 2a but there is no reaction rim formed due to the interaction of tar (T) and semicoke (SC). Vitrinite (V) also present showing desiccation cracks.

Often the cavities and cracks or surfaces of semicoke and coke fragments are lined by tar, indicating the passage of tar (Plate 3a-b). Tar within the pre-carbonization stage towards the base of the wastepile is often isotropic, soft and appears to have reacted with the coal fragments (Plate 4a). This tar is relatively hot and possibly acted not only as a binder but also as a hydrogen donor to vitrinite particles. The briquetting of angular inertodetrinite fragments by tar is shown in Plate 4b. There is no apparent reaction between tar and inertodetrinite.

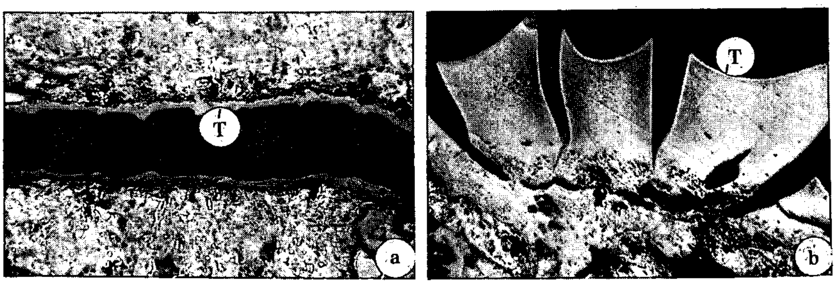

PLATE 3 Conditions same as Plate 1.

a) The presence of thin lining of isotropic tar (T) indicating the passage of liquids and gases through semicoke fragments; b) The deposition of tar (T) in the cavity has lead to the formation of flower-like structures.

The morphology of the vitrinite fragments being briquetted by tar may give an indication as to the temperature of combustion. Usually rounded vitrinite fragments, as shown in Plate 4c, are indicative of higher temperatures as opposed to angular vitrinite fragments (Plate 4d) which indicate lower temperatures.

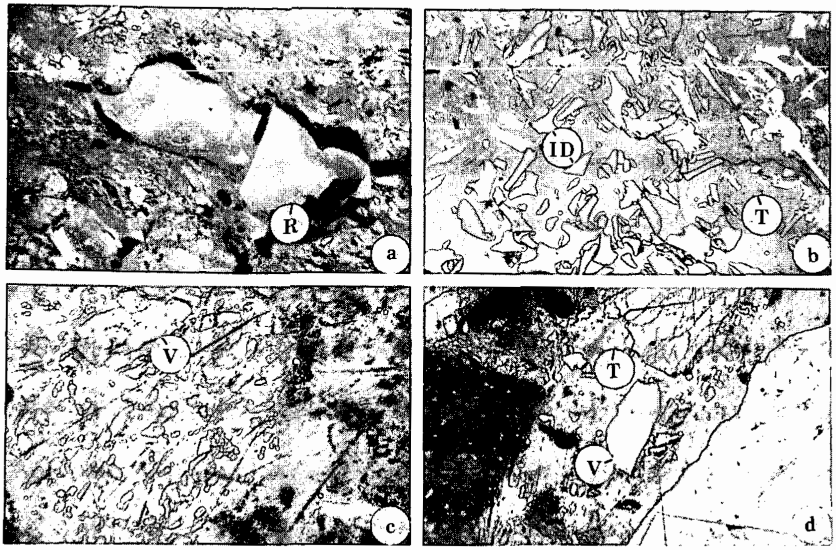

PLATE 4 Conditions same as Plate 1.

a) Tar reacting with coal fragments in pre-carbonization stage to form distinct reaction rims (R). The tar is hot and not only acts as a binder to briquette coal fragments but also as a hydrogen donor; b) Tar (T) is shown here briquetting angular inertodetrinite (ID); c) Tar briquetting rounded to semirounded vitrinite (V); d) Angular vitrinite (V) and inertinite being briquetted by tar (T).

CONCLUSION

The following conclusion can be drawn:

1) Tar generated from the combustion of coal may interact with coal and semicoke/coke fragments. Tar may not only act as a binder but also as a hydrogen donor to coal fragments, thus forming a distinct reaction rim.

2) Different tar types (primary and secondary) can be identified based on morphology, optical texture and reflectance.

3) Tar will briquette loose fragments of coal, as in the case of a wastepile but when coal is present in the form of a coherent seam, briquetting does

not take place but the coal is transformed into carbonized residue impregnated by tar.

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